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CONTROL OF A CATALYTIC FLOW REVERSAL REACTOR MODEL BY LINEAR QUADRATIC REGULATOR

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Abstract: A linear infinite dimensional state space representation of a catalytic flow reversal reactor is used to formulate a state LQ-feedback operator via the solution of a Riccati differential equation. Flow velocity is used to keep the temperature and mole fraction of reactant in the reactor moving along a desired stationary state. Numerical simulations are used to show the performance of the formulated controller.

Keywords: Linear Quadratic Regulator; Infinite Dimensional System; Flow Reversal Reactor

1. INTRODUCTION

Catalytic flow reversal reactors (CFRR) are fixed-bed reactors with periodic reversal of the flow direction. The primary advantage of the technology is that the thermal capacity of the solid material within the reactor acts as a regenerative heat exchanger, allowing autothermal operation without the use of additional energy supply. Catalytic flow reversal reactors have received much attention in recent years (Matros and Bunimovich, 1996) and have been used for many reacting systems, including oxidation of volatile organic compounds (VOCs), oxidation of sulphur dioxide (SO2) and methane combustion.

For exothermic reactions, reversing the flow direction periodically creates a heat trap effect. This effect can be used to achieve and maintain an enhanced reactor temperature compared to a single flow direction mode of operation.

To control the temperature and conversion in a CFRR unit, Budman *et al.* (1996) used a feedback PID and a feedforward control strategy, both with set points obtained from operability map (parametric study). A model predictive control scheme was presented by Dufour and Toure (2004) for the combustion of volatile organic compounds in a CFRR unit.

Linear-Quadratic (LQ) optimal control has been used to control flow reversal reactors in Edouard *et al.* (2005). In Eduard *et al.*, a LQ controller was formulated and applied to a CFRR unit with fast frequency of flow reversal. For this specific case of operation, Eduard *et al.* took advantage of the high frequency of flow reversal and approximated the mathematical model of the reactor by a countercurrent reactor system. A LQ controller was obtained from a linear model derived by linearization of the countercurrent model at a constant operating condition and a finite difference discretization of the resulting model.

In this paper, a linear-quadratic optimal controller is formulated for a catalytic flow reversal reactor using an infinite dimensional Hilbert space rep-

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resentation of the system. The LQ controller is formulated to keep the distribution of the temperature and mole fraction of reactant along the reactor at stationary state by using the flow velocity. A linear infinite dimensional state space description is used to formulate the controller. Using the infinite dimensional state space, a state LQ-feedback operator is computed via the solution of a Riccati differential equation.

Our motivation to select an LQ controller was based on the fact that LQ control is considered key technique due to the fact that it provides an optimal solution with respect to a clearly defined objective. In addition, LQ control has been largely studied for infinite dimensional systems. Our objective was therefore to analyze the performance of such a reference controller in a process example (catalytic reactor) and provide results that can be used to evaluate its performance and possibly its limitations.

2. PRINCIPLE OF OPERATION

The principle of the heat trap effect in catalytic flow-reversal reactors is illustrated in Figure 1. Figure 1(a) illustrates a reactor temperature profile that might be observed in a standard unidirectional flow operation for a combustion. If a temperature pattern, shown in Figure 1(a) and (b) is established, the reverse flow operation can then be used to take advantage of the high temperatures near the reactor exit to pre-heat the reactor feed. A quasi-steady state operation may be achieved in which the reactor temperature profile has a maximum value near the centre of the reactor, which slowly oscillates as the feed is periodically switched between the two ends of the reactor, as shown in Figure 1(c-e). The quasi-steady state operation will be referred in this paper as *stationary state*.

3. MODEL DESCRIPTION

The dynamic of the catalytic flow reversal reactor can be described by partial differential equations (PDE's) derived from mass and energy balances. A wide range of model with different degrees of complexities have been proposed in the literature for catalytic flow reversal reactors for different reacting systems. One of the most complete models is given in Salomons *et al.* (2004), where a 2-dimensional dynamic heterogenous convection-diffusion-reaction model is used to simulate the effect of different operating conditions on the combustion of lean fugitive emissions. In this work, a simplified model is used where plug flow is assumed and a single average value for the states

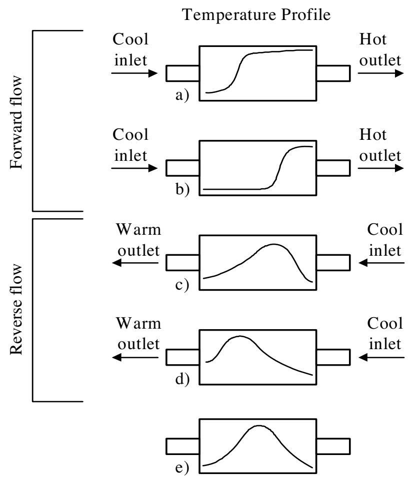

Fig. 1. Illustration of the heat trap effect for reverse flow operation.

variables in the fluid and solid phase is used (pseudo-homogeneous model).

3.1 Mathematical model:

The basic equations for the mass and energy balances for a pseudo-homogeneous plug-flow model are:

$$\epsilon \frac{\partial Y}{\partial t} + \alpha v_s \frac{\partial Y}{\partial z} = -k_0 e^{\frac{-E}{R_g T}} Y \quad (1)$$

$$\eta \frac{\partial T}{\partial t} + \alpha v_s \rho \frac{\partial T}{\partial z} = (-\Delta H_r) k_0 C e^{\frac{-E}{R_g T}} Y \quad (2)$$

where the parameters k_0, η and ρ are given by $k_0 = (1-\epsilon)\mu_{eff} k_\infty, \eta = \rho_s(1-\epsilon)Cp_s$ and $\rho = \rho_f Cp_f$, and with the boundary conditions given, for $t \geq 0$, by:

$$Y(0,t) = Y_{in} \quad \text{and} \quad T(0,t) = T_{in}. \quad (3)$$

Y and T are the average mole fraction of methane and temperature, respectively; v_s is the superficial velocity, α is the fraction of mass flow velocity (used as manipulated variable) and ϵ is the bed porosity. Values for the model parameters are given in Table 1. The initial conditions are given, for $0 \leq z \leq 1$ by:

$$Y(z,0) = Y_0(z) \quad \text{and} \quad T(z,0) = T_0(z). \quad (4)$$

The fluid in the reactor is treated as an ideal gas: $\rho_f = \frac{P}{R_g T}$ and $C = \frac{P}{R_g T}$. We neglect any pressure drop in the reactor and we consider that density is constant at conditions equal to the inlet conditions. These considerations result in $v_s = v_{in}$ (Salomons *et al.*, 2004).

3.2 Dimensionless Model

In this subsection the dynamics of the catalytic flow-reversal reactor will be described by means of an infinite-dimensional system description derived from an equivalent nonlinear PDE dimensionless model. Such an approach is standard in tubular reactor models, see e.g. (Aksikas *et al.*, 2007). Let us consider the following state transformation:

$$\theta_1 = \frac{Y_{in} - Y}{Y_{in}}, \quad \theta_2 = \frac{T - T_{in}}{T_{in}} \tag{5}$$

Then we obtain the following equivalent representation of the model (1)-(3):

$$\frac{\partial \theta_1}{\partial t} = \alpha v_1 \frac{\partial \theta_1}{\partial z} + k_1(1-\theta_1)\exp(\frac{\mu\theta_2}{1+\theta_2}) \tag{6}$$

$$\frac{\partial \theta_2}{\partial t} = \alpha v_2 \frac{\partial \theta_2}{\partial z} + k_2(1-\theta_1)\exp(\frac{\mu\theta_2}{1+\theta_2}) \tag{7}$$

where the dimensionless v_1, v_2, ν, k_1 and k_2 are related to the original parameters as follows:

$$v_1 = -\frac{v_{in}}{\epsilon}, \quad v_2 = -\frac{v_{in}\rho T_{in}}{\eta}, \quad \mu = \frac{E}{R_g T_{in}},$$
$$k_1 = \frac{k_0}{\epsilon}\exp(-\mu), \; k_2 = \frac{(-\Delta H_r)Ck_1\epsilon}{\eta}$$

3.3 Infinite Dimensional Linearized Model

Let us denote by $\theta_e := (\theta_{1,e}, \theta_{2,e})$ and α_e the dimensionless equilibrium profile of the model (6-7). Let us consider the state transformation:

$$x(t) := \begin{bmatrix} x_1(t) \\ x_2(t) \end{bmatrix} := \begin{bmatrix} \theta_1(t) - \theta_{1,e} \\ \theta_2(t) - \theta_{2,e} \end{bmatrix}, \tag{8}$$

with the new input vector $u(t) := \alpha(t) - \alpha_e$. Then the linearization of the system (6)-(7) around its equilibrium profile leads to the following linear infinite-dimensional system on the Hilbert space H:

$$\begin{cases} \dot{x}(t) \; = Ax(t) + Bu(t) \\ x(0) \; = x_0 \in H \; . \end{cases} \tag{9}$$

Here A is the linear operator defined on its domain:

$$D(A) = \{x : x \text{ is a.c.}, \; \frac{dx}{dz} \in H \text{ and } x(0) = 0\}, \tag{10}$$

by

$$Ax = \begin{bmatrix} \alpha_1 \frac{d.}{dz} + \beta_1 I & \beta_2 I \\ \beta_3 I & \alpha_2 \frac{d.}{dz} + \beta_4 I \end{bmatrix} \begin{bmatrix} x_1 \\ x_2 \end{bmatrix}, \tag{11}$$

where the functions α_i and β_i are given by

$$\alpha_i(z) = v_i \alpha_e(z), \; i = 1, 2,$$
$$\beta_1(z) = -k_1 \exp(\frac{\mu\theta_{2,e}}{1+\theta_{2,e}}),$$
$$\beta_2(z) = \frac{\mu k_1(1-\theta_{1,e})}{(1+\theta_{2,e})^2}\exp(\frac{\mu\theta_{2,e}}{1+\theta_{2,e}}),$$
$$\beta_3(z) = -k_2 \exp(\frac{\mu\theta_{2,e}}{1+\theta_{2,e}}),$$
$$\beta_4(z) = \frac{k_2(1-\theta_{1,e})}{(1+\theta_{1,e})^2}\exp(\frac{\mu\theta_{2,e}}{1+\theta_{2,e}}).$$

The operator $B \in \mathcal{L}(L^2(0,1), H)$ is the linear bounded operator given by

$$B = \begin{bmatrix} \gamma_1 I \\ \gamma_2 I \end{bmatrix}, \tag{12}$$

where the functions γ_i are given by

$$\gamma_i(z) = v_i \frac{d\theta_{i,e}}{dz}, \; i = 1, 2.$$

The following proposition deals with the exponential stability of the linearized catalytic flow-reversal model. This property will play an important role in LQ-control design, especially the existence and the uniqueness of a solution (see Lemma 2 below).

Proposition 1. Let us consider the operator A defined by (10)-(11). Then A generates an exponentially stable C_0-semigroup on H.

Proof: By using the fact that the functions $\alpha_i, \; i = 1, 2$ are negative (since the constants $v_i, i = 1, 2$ are negative), then it can be shown that the eigenvalues of the operator A are of the following form, see (Christofides, 2001, p. 18):

$$\lambda_A = -\infty + n\pi i, \; n = -\infty, \ldots, \infty.$$

4. CONTROLLER DESIGN

In this section, we are interested in the linear-quadratic optimal (LQ) problem (see e.g. (Curtain and Zwart, 1995) and references therein) in order to design a state LQ-optimal controller for the linearized catalytic flow-reversal reactor described by (9)-(12). First let us define an output function $y(\cdot)$ by

$$y(t) = Cx(t) := \begin{bmatrix} w_1 I \; w_2 I \end{bmatrix} x(t), \; t \geq 0, \tag{13}$$

where $w_1, w_2 : [0,1] \rightarrow I\!R$ are continuous functions. Now let us consider the LQ-optimal control problem: for any initial state $x_0 \in H$, find a square integrable control $u_{opt} \in L^2[0, \infty; L^2(0,1)]$ which minimizes the cost functional

$$J(x_0, u) = \int_0^\infty (\langle Cx(t), Cx(t)\rangle + \langle u(t), Ru(t)\rangle)dt, \tag{14}$$

where R is a coercive function. The solution of this problem can be obtained by finding the positive self-adjoint operator $Q_o \in \mathcal{L}(H)$ which solves the operator Riccati equation, viz

$$[A^*Q_o + Q_oA + C^*C - Q_oBR^{-1}B^*Q_o]x = 0, \quad (15)$$

for all $x \in D(A)$, where $Q_o(D(A)) \subset D(A^*)$.

It is known that under the exponential stabilizability of (A, B) and the exponential detectability of (C, A), the operator Riccati equation admets a unique positive self-adjoint solution: see e.g. (Curtain and Zwart, 1995, Section 6.2). Then the following lemma is an immediate consequence of Proposition 1.

Lemma 2. Consider the linearized catalytic flow-reversal reactor model (9)-(11), with control operator B given by (12) and observation operator C given by (13). Then the operator Riccati equation (15) has a unique positive self-adjoint solution $Q_o \in \mathcal{L}(H)$ and for any initial state $x_0 \in H$, the quadratic cost (14) is minimized by the unique control u_{opt} given on $t \geq 0$ by

$$u_{opt}(t) = K_o x(t)\ , x(t) = e^{(A+BK_o)t}x_0\ , \quad (16)$$

where the optimal feedback

$$K_o = -R^{-1}B^*Q_o \in \mathcal{L}(H, L^2(0,1)) \quad (17)$$

is *stabilizing*, i.e. the feedback C_0-semigroup $(e^{(A+BK_o)t})_{t\geq 0}$ is exponentially stable. In addition, the optimal cost is given by $J(x_0, u_{opt}) = \langle x_0, Q_o x_0\rangle$.

Now we are in a position to establish the following theorem:

Theorem 3. Let us consider the linearized catalytic flow-reversal reactor model (9)-(11), with control operator B given by (12) and observation operator C given by (13). Let ψ_1, ψ_2 be the solutions of the system equations:

$$\begin{cases} 2\beta_1\psi_1 + w_1^2 - \gamma_1^2 r^{-1}\psi_1^2 = \dfrac{d(\alpha_1\psi_1)}{dz}, \\ 2\beta_4\psi_2 + w_2^2 - \gamma_2^2 r^{-1}\psi_2^2 = \dfrac{d(\alpha_1\psi_2)}{dz}, \\ \beta_3\psi_2 + \beta_2\psi_1 + w_1 w_2 = \gamma_1\gamma_2 r^{-1}\psi_1\psi_2, \\ \psi_1(1) = 0, \\ \psi_2(1) = 0. \end{cases} \quad (18)$$

Then the optimal LQ-feedback oprator is given for all $x \in H$ by

$$K_o x = -\gamma_1\psi_1 x_1 - \gamma_2\psi_2 x_2. \quad (19)$$

Proof: In view of the structure of the operator A and in order to solve the operator Riccati equation (15), it seems natural to look for a solution of the form

$$Q_o = \begin{bmatrix} \psi_1 I & 0 \\ 0 & \psi_2 I \end{bmatrix}.$$

On the other hand, it can be shown by straightforward calculation that if the functions ψ_1 and ψ_2 are solutions of the differential equations system (18), then Q_0 is solution of the operator Riccati equation (15).

5. NUMERICAL SIMULATION

To show the effectiveness of the LQ-feedback controller, the formulated controller is used for a CFRR unit for combustion of methane emissions. CFRR technology has been suggested for the combustion of lean methane streams for reduction of greenhouse gases emissions from natural gas transmission facilities, upstream oil and gas production facilities and coal beds (Hayes, 2004).

To simulate the CFRR operation, we use a 1-dimensional pseudo-homogeneous model as given in equation (1)-(2). Model parameters were taken from Salomons *et al.* (2004) and are given in Table 1. All computer simulations were performed in COMSOL multi-physics environment. A scheme of the reactor configuration used in simulations is given in Figure 2.

Table 1. Model Parameters.

Parameter	Value
k_∞	1.35E5 s^{-1}
ϵ	0.51
E	54,400 $kg \cdot m^2 \cdot s^{-2} \cdot mol^{-1}$
R_g	8.314 $kg \cdot m^2 \cdot s^{-2} \cdot mol^{-1} \cdot K^{-1}$
ρ_s	1240 $kg \cdot l^{-1}$
Cp_f	1,066 $J \cdot kg^{-1} \cdot K^{-1}$
Cp_s	1,020 $J \cdot kg^{-1} \cdot K^{-1}$
ΔH_r	-802E3 $J \cdot mol^{-1}$
P	101325 $kg \cdot m^{-1}s^{-2}$
v_{in}	1 $m \cdot s^{-1}$
$\overline{M}$	0.02896 $kg \cdot mol^{-1}$
μ_{eff}	1

Using an arbitrarily chosen nominal operating conditions,

$$Y(0,t) = 0.001,\ T(0,t) = 298K,\ v_{in} = 1\ m \cdot s^{-1}$$

and the pseudo-homogeneous model given in equation (1)-(2), the stationary state is computed for a full cycle time of 600 *sec* (forward + reverse flow), see Figure 3.

An LQ-feedback controller is computed using the linearized model, equation (9). Linearization is performed around a single distribution of the stationary state. It is assumed that the process model does not change much with linearization profile used for a relatively narrow range of flow reversal frequencies. The study of the effect of the linearization profile is subject of future work.

Fig. 2. Ilustration of CFRR unit used for numerical simulations. Arrows indicate inlet/outlet gas flow direction.

The LQ-control law that results from solving the system of equations (18) is given in Figure 4 for $w_1(z) = 1$ and $R(z) = 1$. To evaluate

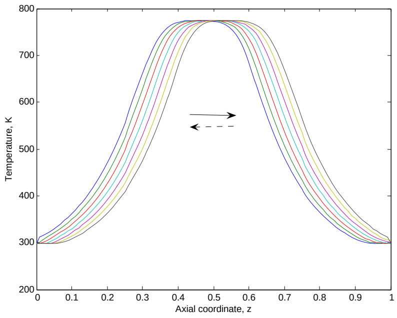

Fig. 3. Temperature distributions at stationary state. Distributions are taken every 50 *sec*.

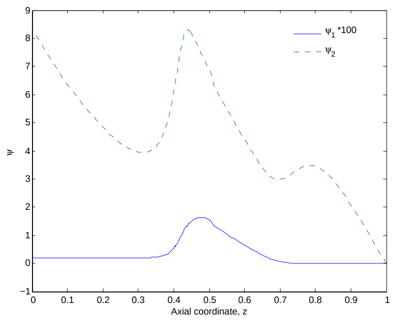

Fig. 4. LQ-feedback functions ψ_i for $w_1(z) = 1$ and $R(z) = 1$.

the performance of the controller, we computed the closed-loop response for a case where the initial state variables are not at stationary state (Figure 5). For the controller calculations, all state variables are assumed to be available and 120 discrete points are used as measuring points. The closed-loop temperature response at three points along the reactor is shown in Figure 6. It can be observed that the state converges quickly to the stationary state. The trajectory of the manipulated variable is shown in Figure 7.

A comparison of the closed-loop response and the open loop response for the reactor system that is started with a distribution of the states that do not correspond to the stationary state is given in Figure 8.

Since it is not practical to manipulate the flow velocity (αv_s) along the axial coordinate, we ap-

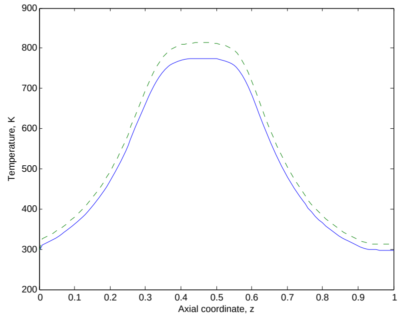

Fig. 5. Initial temperature distribution for closed-loop simulation (dashed line) and temperature distribution at stationary state at the beginning of the forward flow operation (solid line).

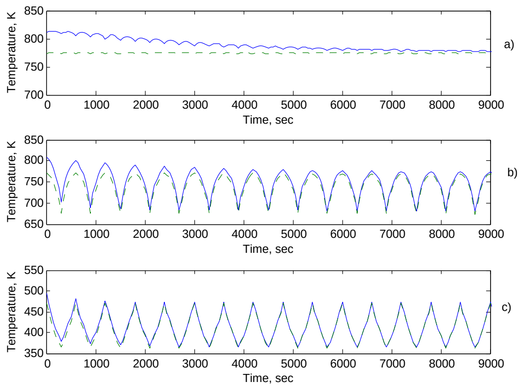

Fig. 6. Closed-loop temperature trajectory (solid line) and temperature trajectory at stationary state (dashed line) for a) $z = 0.2$, b) $z = 0.4$, c)$z = 0.5$.

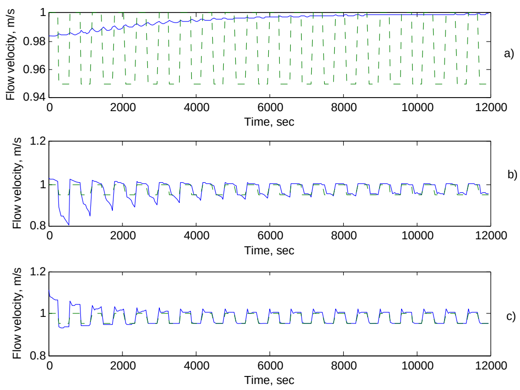

Fig. 7. Closed-loop input variable (solid line) and input variable at stationary state (dashed line) for a) $z = 0.2$, b) $z = 0.4$, c)$z = 0.5$.

proximate the optimal distribution of the manipulated variable by averaging the flow velocity. We consider the case where the flow velocity can be imposed at two spatial points: reactor inlet and mid-section. The velocity for the first half of the reactor, imposed at the reactor inlet, is obtained from averaging the optimal input from $z = 0$ to $z = 0.5$. The velocity for the second half of the reactor, imposed at the mid-section of the reactor,

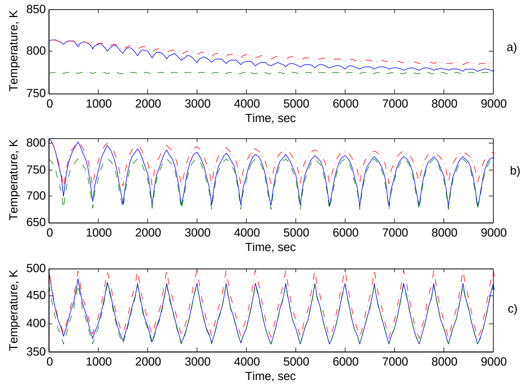

Fig. 8. Closed-loop input variable (solid line), open-loop input variable (dot-dashed line) and input variable at stationary state (dashed line) for a) $z = 0.2$, b) $z = 0.4$, c)$z = 0.5$.

is obtained from averaging the optimal input from $z = 0.5$ to $z = 1$. The closed-loop temperature response at three points along the reactor is shown in Figure 9. The trajectory of the input variable at the inlet and midsection of the reactor are shown in Figure 10.

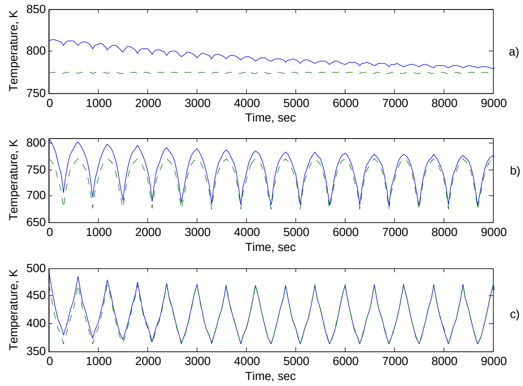

Fig. 9. Closed-loop temperature trajectory (solid line) and temperature trajectory at stationary state (dashed line) for a) $z = 0.2$, b) $z = 0.4$, c)$z = 0.5$ (averaged input).

6. CONCLUSIONS

In this paper, we presented the formulation of an LQ-feedback controller for a catalytic flow-reversal reactor. A linear infinite dimensional state-space representation of the system is used to formulate a state LQ-feedback operator via the solution of the corresponding Riccati algebraic equation. The controller is aimed at keeping the state variables, temperature and concentration, at the stationary state. Flow velocity is used to control the system. Numerical simulations are used to show the response of the closed loop system.

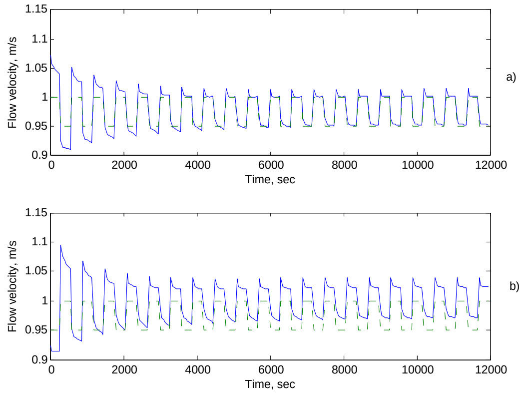

Fig. 10. Closed-loop input variable (solid line) and input variable at stationary state (dashed line) for a) $z = 0$ and b) $z = 0.5$ (averaged input).

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